

## CLAIMS

1. A lithium ion secondary battery comprising a positive electrode capable of absorbing and desorbing lithium ions and a negative electrode including a first layer that is largely composed of carbon and a second layer that contains an element to be alloyed with lithium, wherein the lithium content of the second layer is between 31 and 67 atomic-% at a discharge depth of 100 %.

2. The lithium ion secondary battery claimed in claim 1, wherein the capacity of the negative electrode is higher than that of the positive electrode.

3. The lithium ion secondary battery claimed in claim 2, wherein lithium in a quantity that satisfies the following formulas (1) and (2) is electrically connected to the positive electrode or the negative electrode:

$$Li = Cb(1 - L_c) + (M_{atom} \times L_s / (1 - L_s)) \times Li_{capa} \cdots (1);$$

$$Li + Cat \leq Cb + M_{atom} \times M_{capa} \cdots (2);$$

(wherein Li represents the capacity of Li electrically connected to the positive electrode or the negative electrode, Cb represents the capacity of active material contained in the first layer, L<sub>c</sub> represents the initial charging/ discharging efficiency of the first layer, M<sub>atom</sub> represents the number of atoms of lithium absorbing material contained in the second layer, L<sub>s</sub> represents the Li content in the second layer at a discharge depth of 100 %, Li<sub>capa</sub> represents the capacity of a lithium atom, Cat represents the capacity of the positive electrode, and M<sub>capa</sub> represents the capacity of an atom of lithium absorbing material contained in the second layer).

4. The lithium ion secondary battery claimed in one of claims 1 to 3, wherein the element to be alloyed with lithium is at least one selected

from Si, Ge, In, Sn, Ag, Al and Pb.

5. The lithium ion secondary battery claimed in one of claims 1 to 4, wherein, as the element to be alloyed with lithium, Si and/ or Sn is/ are included.

6. The lithium ion secondary battery claimed in one of claims 1 to 5, wherein the first layer includes at least one selected from graphite, fullerene, carbon nanotube, diamond like carbon, amorphous carbon, and hard carbon.

7. The lithium ion secondary battery claimed in one of claims 1 to 6, wherein the active material of the positive electrode includes at least one compound selected from lithium cobalt oxide, lithium manganese oxide, and lithium nickel oxide.

8. The lithium ion secondary battery claimed in one of claims 1 to 7, wherein the active material of the positive electrode includes lithium manganate.

9. A method for using a lithium secondary battery comprising a positive electrode capable of absorbing and desorbing lithium ions and a negative electrode including a first layer that is largely composed of carbon and a second layer that contains an element to be alloyed with lithium, wherein the lithium content in the second layer of the negative electrode is made between 31 and 67 atomic-% on completion of discharge.

10. The method for using a lithium ion secondary battery claimed in claim 9, wherein the capacity of the negative electrode is higher than that of the positive electrode.

11. The method for using a lithium ion secondary battery claimed in claim 9 or 10, wherein the element to be alloyed with lithium is at least one selected from Si, Ge, In, Sn, Ag, Al and Pb.

12. The method for using a lithium ion secondary battery claimed in one of claims 9 to 11, wherein, as the element to be alloyed with lithium, Si and/ or Sn is/ are included.

13. A method for manufacturing a lithium ion secondary battery comprising a positive electrode capable of absorbing and desorbing lithium ions and a negative electrode, involving the step of, after forming the negative electrode including a first layer that is largely composed of carbon and a second layer that contains an element to be alloyed with lithium, adding lithium in a quantity that satisfies the following formulas (A) to (D) to the surface of the positive electrode or the negative electrode:

$$Cb + M_{\text{atom}} \times M_{\text{capa}} > Cat \cdots (A);$$

$$0.31 \leq L_s \leq 0.67 \cdots (B);$$

$$Li = Cb (1 - L_c) + (M_{\text{atom}} \times L_s / (1 - L_s)) \times Li_{\text{capa}} \cdots (C);$$

$$Li + Cat \leq Cb + M_{\text{atom}} \times M_{\text{capa}} \cdots (D);$$

(wherein Li represents the capacity of Li electrically connected to the positive electrode or the negative electrode, Cb represents the capacity of active material contained in the first layer of the negative electrode,  $L_c$  represents the initial charge/ discharge efficiency of the first layer of the negative electrode,  $M_{\text{atom}}$  represents the number of atoms of lithium absorbing material being active material contained in the second layer of the negative electrode,  $L_s$  represents the Li content in the second layer of the negative electrode at a discharge depth of 100 %,  $Li_{\text{capa}}$  represents the capacity of a lithium atom, Cat represents the capacity of the positive

electrode, and  $M_{\text{capa}}$  represents the capacity of an atom of lithium absorbing material being active material contained in the second layer of the negative electrode).